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Research Article

Generation of Coherent XUV Radiation in Gas Mixture of N_2 and H_2 using High Power Laser System

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Abstract

High harmonic generation is studied in gas species, namely pure H_2, pure N_2, and their mixture in 1:1 ratio. Observation of harmonic orders from 17H to 35H orders is well resolved. The harmonic spectrum produced from pure H_2 is obtained, and the harmonic signal is weaker compared to that produced from pure N_2 and their mixture. On the other hand, harmonic yield in the gas mixture is enhanced compared to that from pure H_2. The maximum photon energy that harmonic orders can reach is calculated with the well-known cutoff formula. The experiment and the calculation results are in close agreement. In the mixture of N_2:H_2, the higher harmonics are enhanced compared to the plateau region harmonics. The enhancement of 35H orders in pure N_2 or H_2-N_2 mixture is observed compared to pure H_2.

Keywords: Atomic and molecular physics, gas mixture, femtosecond laser, high harmonic generation, extreme ultraviolet (XUV)

Yüksek Güçlü Lazer Sistemi Kullanılarak N_2 ve H_2 Gaz Karışımında Koherent XUV Radyasyonunun Üretilmesi

Öz

Saf H_2, saf N_2 ve bunların 1:1 oranında karışımları yüksek harmonik üretiminde çalışılmıştır. 17H'den 35H'ye kadar olan harmonik mertebeler gözlemlendi. Saf H_2'den üretilen harmonik spektrum elde edildi, ancak harmonik sinyal, saf N_2 ve bunların karışımından üretilen harmonik spektruma kıyasla daha zayıftır. Gaz karışımındaki harmonik verim, saf H_2'den gelen harmoniklere kıyasla artar. Harmonik mertebelerin ulaşabileceği maksimum foton enerjisi, iyi bilinen cutoff formülü ile hesaplandı. Deney ve hesaplama sonuçlarıyla elde edilen harmonik mertebeler bir uyum içindedir. N_2:H_2 karışımında, plato bölgesi harmoniklerine kıyasla daha yüksek harmonikler geliştirilmiştir. Saf N_2 veya H_2-N_2 karışımında 35H mertebelerinde saf H_2'ye göre artış gözlendi.

Anahtar Kelimeler: Atomik ve moleküler fiziği, gaz karışımı, femtosaniye lazer, yüksek harmonik üretimi, aşırı ultraviyole (XUV).

1. Introduction

High-order harmonics generation (HHG) is a way to generate a coherent powerful extreme ultraviolet (XUV) radiation source (Chang and Corkum, 2010; Kapteyn et al., 2005). This coherent XUV source has potential for many practical applications, such as the study of the atomic and molecular process (Remetter et al., 2006), quantum dynamics of the electronic wave packet (Paul et al., 2001), biological imaging (Eisebitt et al., 2004; Itatani et al., 2004; Ravasio

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et al., 2009; Seaberg et al., 2011), production of isolated attosecond laser pulses (Hentschel et al., 2001; Lewenstein, 2002), and so on. The wide range of applications motivates the development of high harmonic generation. The improvement of HHG has opened the realization of extreme nonlinear optics (Popmintchev et al., 2010), quantum interference effect (Vozzi et al., 2006), and molecular self-probing (Haessler et al., 2011). However, the bottleneck of this powerful light source is its low conversion energy, i.e. conversion from fundamental field to its higher frequencies. There are many experimental and theoretical studies to increase the efficiency of HHG in pure atomic and molecular gases (Brizuela et al., 2013; Ishikawa, 2003; Kanai et al., 2007; Takahashi et al., 2007). However, there are still many open questions to optimize the efficiency of the high harmonics because HHG strongly depends on macroscopic conditions such as phase matching, which can vary for different experimental approaches and designs.

Microscopic physics of harmonic generation can be well explained with a three-step model (Corkum, 1993; Lewenstein et al., 1994). In this model, an electron exposed to an intense laser field tunnels through the atomic barrier, and then the electron is driven back and forth with the oscillating laser field. In this process, the electron gains kinetic energy. Finally, the electron could recombine with its core and releases its kinetic energy in the form of high-energy photons. The highest harmonic energy (cutoff) is given by $E_{\max} = I_P + 3.17U_p$ (Lewenstein et al. 1994), where I_p is the ionization potential of the used gas, $U_p \sim I\lambda^2$ is the ponderomotive energy gained by the free electron due to the laser oscillation, and it is proportional to the laser intensity (I) and wavelength (λ) (Corkum, 1993; Lewenstein et al. 1994). There are several parameters to control the efficiency of HHs and to extend the cutoff harmonics. For example, the highest harmonic energy is proportional to the laser intensity (I) and the wavelength (λ). By controlling these two variables, one can optimize the harmonic signal and can extend the cutoff harmonic. On the other hand, there is a limitation to increase laser intensity (~10^{17}W/cm^2) because the magnetic component of the laser field is comparable to the electric field of the pulse.

In this paper, we report harmonic generation from pure N_2, pure H_2, and their mixture using the high power laser system producing optical pulses having energy per pulse 6mJ with 50fs pulse duration at a 10 Hz repetition rate. The highest harmonics from 31H to 35H in the mixed gas medium are enhanced as a factor of ~3 and ~4 according to H_2, (high conversion efficiency). The results could be explained as that the harmonics produced in N_2 medium boost harmonics produced in pure H_2. The effect of the gas mixture has the potential to increase or decrease the efficiency of the HHG. Ionization potential (I_p) of both H_2 and N_2 are close to each other (I_p of N_2 molecules is 15.7eV (Tate and Smith, 1932)) and I_p of H_2 molecule is 15.4eV (Bleakney, 1932)), and they present well-resolved harmonic peaks. Therefore, the H_2-N_2 gas mixture is chosen for investigating the characteristic of harmonic generation.

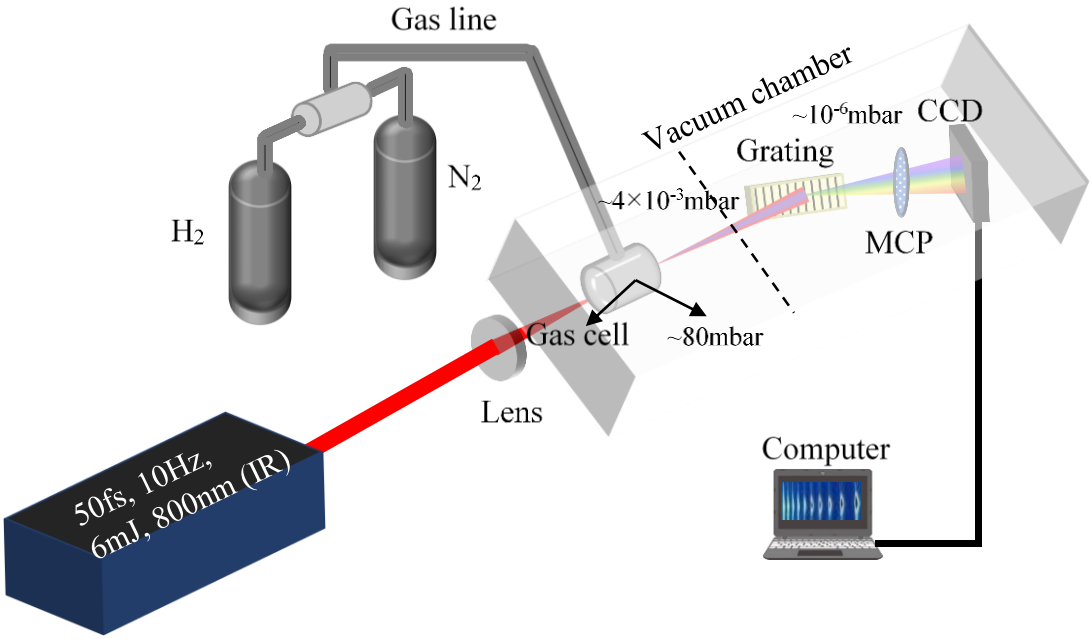

Figure 1. Schematic of the harmonic generation experimental setup. The setup includes a gas cell (harmonic source), a grating (for dispersion of harmonic spectrum), MCP (to amplify the harmonic yield), CCD (to capture the harmonics signal), IR (infrared radiation)

2. Experimental Setup

The laser system has been used that the driving laser field consists of high power laser system producing optical pulses about a 10Hz repetition rate, 50fs pulse duration (~19cycle) centered at 800nm, and energy of 6mJ per pulse. To generate high-order harmonics, the radiation is focused on a gas cell, which is machined in a machine shop to generate high harmonics. The gas cell has a length of 7mm and a diameter of 5mm. The cell has input and output holes, which are taped by aluminum foil tape to localize gas density inside the cell. The strong laser field is focused on the gas cell by using a 40cm focal length lens, and high-order harmonics of the fundamental field are generated. The intensity at the interaction region is estimated at $1\times10^{15}Wcm^{-2}$. After the gas cell, the generated harmonics propagate through to the spectrometer, and they are recorded. The schematic of the experimental design is presented in Fig. 1. The recorded data are analyzed by using the MATLAB program.

The powerful laser system allows one to control harmonic orders, and it can reach a high cutoff harmonics due to its high power. The generated harmonics propagate through the spectrometer. First, each harmonic order is reflected from the grating at a different angle. Second, the harmonics are amplified by a microchannel plate (MCP), and they are finally captured by the charge-coupled device (CCD) camera. The state-of-the-art laser system allows reaching higher-order harmonics by enhancing the cutoff harmonic. The cutoff harmonic in the mixed gases is decreased compared to that in pure N_2. The results can be explained as the harmonics coming from N_2 are absorbed by the H_2. Moreover, destructive interference of both species leads to this decrement. In addition, the ionization potential of both gases is close to each other, and so the phase mismatch induced by the free electron could cause this decrease in the mixture. However, the harmonic yield in the gas mixture is boosted compared to that produced in pure H_2. This mechanism could be constructive interference between harmonics produced in N_2 and H_2 gas species, respectively (Kanai et al., 2007).

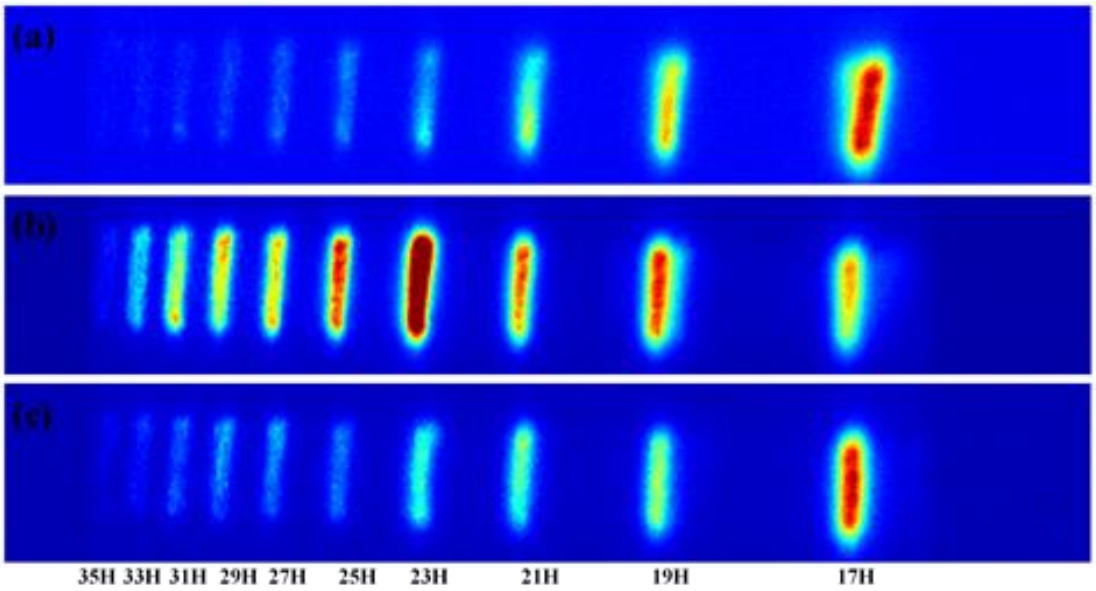

Figure 2. Captured Harmonic images by a CCD camera at the cell pressure of 80mbar. (a) pure H_2, (b) pure N_2 (c) N_2-H_2 mixture in 1:1 ratio

3. Experimental Results

Harmonic spectra are produced in three different gas mediums, N_2, H_2, and their mixture N_2-H_2, respectively. Figure 2 presents the typical CCD image of harmonic spectrum in (a) pure H_2, (b) pure N_2, and (c) N_2-H_2 mixture (50%-50% ratio) at the pressure of 80mbar. Harmonic orders are produced up to 35th harmonic orders, Fig. 2. The interaction chamber pressure is 4×10^{-3}mbar, which corresponds to interaction region pressure, ~80mbar. The integration time of the CCD camera is set to 10s. The fundamental laser field power is 60mW corresponding $\sim1\times10^{15}$Wcm^{-2} focused intensity.

The above-mentioned cutoff formula calculates the maximum photon energy of generated harmonics. The cutoff harmonic order is calculated as the 33rd harmonic order for N_2 and H_2 gas species at $\sim1\times10^{15}$Wcm^{-2} IR laser intensity because the ionization potentials of these two species are close to each other. However, the harmonic signal in pure N_2 is stronger than that in pure H_2. This is attributed to electron dispersion and laser defocusing due to free electron distribution in the medium (Kanai et al., 2007). In addition, the generation of high harmonics in N_2-H_2 gas mixture produces a weak harmonic signal compared to the harmonic signal in pure N_2. This could be due to the absorption of harmonics (produced in N_2) by the H_2 molecules. There is another possibility that there is bright harmonics radiation since the ionization potentials of both species are close and lower. However, the low ionization potential of the medium results in serious electron dispersion and ionization-induced laser defocusing. These drawbacks result in a decrease of the harmonic signal in the gas mixture (Kanai et al., 2007).

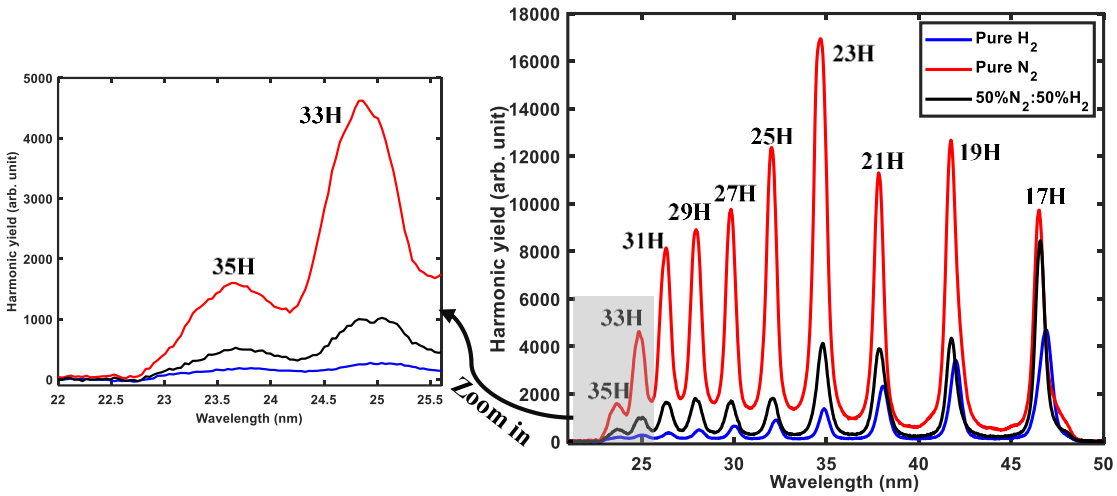

Figure 3. Harmonic spectrum in H_2, N_2, and their mixture with 50%:50% ratio. The solid blue line is for H_2. The solid red line is for N_2. The solid black line is for the mixture. Zoomed in part is for the harmonic spectrum for 33H and 35H orders

Figure 3 is for harmonic yield as a function of wavelength (nm) and photon energy (eV). The solid blue line is for H_2, the solid red line is for N_2, and the solid black line is for the gas mixture. The harmonics are well resolved up to 35 order in all gas species. The gas mixture produces a strong harmonic signal compared to the harmonic signal in pure H_2, Fig. 3. The generated strong harmonics in N_2 gas and the fundamental field (IR) lead to ionization of the H_2 gas in N_2-H_2 gas mixture. The mechanism can lead to an increase of the harmonic signal in N_2-H_2 gas mixture compared to the harmonic signal in pure H_2. Zoomed part of Figure 3 shows the 35^{th} harmonic signal in pure N_2 and the mixture of N_2-H_2.

The decrease of the harmonic signal in the gas mixture of N_2-H_2 compared to the harmonic spectrum in pure N_2 can be explained because of the absorption of the harmonics in the gas mixtures. The absorption in the harmonic spectrum leads to a decrease of the harmonic signal. This paper is an experimental study to increase or decrease the high harmonic signal in the gas mixture. Figure 4 shows harmonic yield as a function of harmonic orders. Figures 4 (a) and (b) present a well harmonic distribution, plateau (19H-31H), and cutoff region (35H). Figure 4 (c) is for the ratio of harmonic yield in the mixture to harmonic yield in pure H_2. The harmonic yield in N_2-H_2 gas mixture is increased (about a factor of ~2 to 4) compared to the harmonic signal in pure H_2, Fig. 4(c). The 35H order produced in pure H_2 is relatively weak compared to other cases. In addition, harmonics near the cutoff region (31H to 35H) are more enhanced compared to those in the plateau region (19H-29H) for the H_2:N_2 gas mixture, Fig. 4(c)).

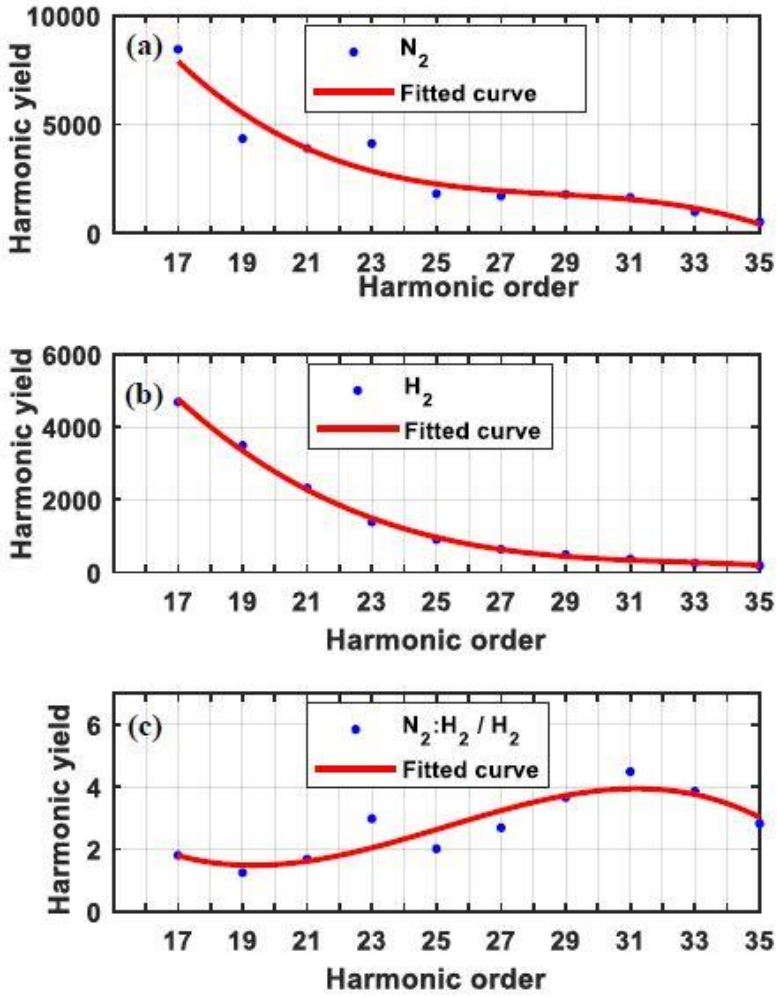

Figure 4. Harmonic yield as a function of harmonic orders. (a) N_2 harmonic yield (b) H_2 harmonic yield (c) N_2:H_2/ H_2 harmonic yield. Blue dots are for experimental measurement, and the solid red line is for fitting

The curve fitting value for Fig. 4 is best performed by using a third-degree polynomial. The function of the fitting is $f(x)=P_1x^3+P_2x^2+P_3x+P_4$, where coefficients ($P_1$, P_2, P_3, and P_4) are given in Table 1. Table 1 presents the harmonic yield polynomial fit for three different gas species. R-square takes values between 0 and 1, and it measures how successful the fit is performed. The value closer to 1 indicates the goodness of the fit. Polynomial coefficients and R square values can be seen in Table 1

Table 1. Fitting function coefficient for Figure 4.

Fitting Function: $f(x)=P_1x^3+P_2x^2+P_3x+P_4$					
Gas Species	**P_1**	**P_2**	**P_3**	**P_4**	**R-square**
N_2	-719.2	940.1	-926.5	2084	0.9283
H_2	-1.079	105.3	-3451	3.833×10^4	0.9977
(N_2: H_2) / H_2	-0.002944	0.2231	-5.323	42.28	0.802

4. Conclusion

Generation of high harmonics extending to ~54eV photon energy is experimentally produced in pure N_2, pure H_2, and their mixture in 1:1 ratio. The focused laser intensity is 1×10^{15}Wcm^{-2} on the gas cell. A high harmonic spectrum of up to 35H orders is generated. The generation of the 35H is increased in the gas mixture compared to the harmonic signal produced in pure H_2 gas. Harmonic spectrums in N_2, H_2, and N_2-H_2 mixture are well resolved from 17H to 35H. The harmonic signal is increased in the gas mixture compared to harmonic yield in pure gas species of H_2 (Fig. 4). The mechanism of harmonic spectrum in the gas mixture can be explained that the harmonics produced in one gas species (N_2) with the fundamental field (IR) result in to increase in the harmonic signal from another gas species (H_2). The harmonic yield as a function of harmonic orders presents harmonic distribution, Fig. 4. Especially, higher orders are enhanced as a factor of 3 to 4. The fitting of harmonic yield is performed with R–squared value close to 1. The optimized harmonic source can be useful for various application areas such as attosecond light source, nonlinear optic in XUV region, and imaging of small-scale object by using harmonic source.

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